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ULTRAVIOLET EXCIMER LASER-BASED IGNITION OF H_2/AIR AND H_2/O_2 PREMIXED FLOWS

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U.S. ARMY LABORATORY COMMAND

BALLISTIC RESEARCH LABORATORY
ABERDEEN PROVING GROUND, MARYLAND

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13. ABSTRACT (Maximum 200 words)
We have demonstrated very efficient ignition of H_2/air and H_2/O_2 premixed flows at room temperature and atmospheric pressure using the radiation from a focussed ArF excimer laser (193 nm). Incident pulse laser energies of less than 1 mJ for H_2/O_2 and 6 mJ for H_2/air are required for ignition with the minimum occurring for lean mixtures. The ignition is due to efficient formation of microplasmas. Unlike the laser-produced microplasmas that are formed by non-resonant photon absorption processes, the resonant ones are easily controlled with respect to spark energy content and thus could be quite useful as potential igniters for the NASP plane and for other supersonic/hypersonic airbreathing applications. We have also found that the atmospheric absorption of the ArF laser radiation strongly affects the values for the incident laser energy (ILE) that is required for ignition. Finally, we report the efficient ignition of C_2H_2/air by the ArF laser which suggests that this uv laser ignition source may have a wider range of applicability.

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I. INTRODUCTION

Pulsed lasers have been used to produce sparks (breakdown) in gases for nearly two decades. Over the intervening years, the details of the laser microplasma formation process have been extensively studied and are currently fairly well understood.[1-3] One of the applications of these laser-produced sparks has been to ignite reactive gases for minimum ignition energy studies.[4] A problem that was quickly discovered in this work was that the laser sparks exhibited a strong threshold behavior for their formation and that once formed, they were frequently so vigorous that they drove detonable gas mixtures into detonation.

Recently, our laboratory has demonstrated the efficient production of microplasmas in various gases by using tunable ultraviolet lasers whose wavelengths correspond to resonance excitation of the constituent atoms. Specifically, we have observed resonant microplasma formation in flows of oxygen-atom containing molecules such as O_2 and N_2O with the laser set at 226 nm, a wavelength which corresponds to oxygen-atom two-photon excitation.[5-7] Similarly, we have observed resonant laser-produced microplasmas in H_2 flows with the laser set at 243 nm, a hydrogen-atom two-photon excitation wavelength.[7] These uv laser produced microplasmas differ significantly from those formed by non-resonant laser radiation in that they are formed with much lower values of incident laser energy (ILE) required, and also they are controlled much more easily with respect to the amount of laser energy that is deposited into the focal volume, i.e., the sharp threshold for breakdown is not observed. A mechnism for the microplasma formation process has been deduced and involves three sequential steps: (a) the multiphoton photochemical production of substituent atoms (H and O), (b) resonant multiphoton ionization of these atoms to efficiently produce "seed" electrons in the laser focal volume, and (c) microplasma formation in the focal volume through the process of electron multiplication due to cascade ionization and plasma heating via the inverse brehmsstrahlung effect.[6]

We have previously used these resonant microplasmas for the ignition of H_2/O_2 and H_2/N_2O premixed flows.[5-7] We have extended this work to include more practical laser systems, ones that could be possibly used in actual field applications. Specifically, since we are interested in the potential of this new igniter source for the National Aerospace Plane (NASP) applications, we have chosen to work with one of the common uv gas discharge lasers, the ArF excimer laser, which operates at 193 nm. This paper describes not only the results of our ignition studies with the ArF laser, but also the results of studies aimed at the understanding of some of the underlying physical and chemical mechanisms entailed in this phenomenon.

II. EXPERIMENTAL

The experimental apparatus has been described previously.[6] Briefly, a Lumonics excimer laser (Model 440) is focused by a 10 cm focal length lens into a premixed flow of H_2/O_2 or H_2/air at room temperature. This flow passes through the orifice (0.7 mm) of a jet burner and intersects the laser focal volume approximately 1-2 mm above the burner surface. The criterion for ignition is straightforward, i.e., ignition is recorded when a flame appears following the laser pulse. The flow conditions are set so that the laser-generated flame is stabilized on the burner. Following ignition, the flame is

quickly extinguished and the water-cooled burner is allowed to return back to ambient conditions.

For the H_2 microplasma formation experiments, the laser used is a Nd:YAG/pumped dye laser system whose radiation in the wavelength region near 243 nm is generated by frequency doubling the dye laser and mixing this doubled beam with the residual 1.06 micron beam from the Nd:YAG pump laser. Typically for these experiments we operated in the 0.1-1 mJ/pulse range while the system is capable of delivering up to 3 mJ/pulse.

For the oxygen-atom spin-orbit studies, the Nd:YAG/dye laser was operated at 226 nm (O-atom two-photon transition) with low pulse energies used (0.5 mJ or less) and a long focal length lens (f.l. = 40 cm) so as to avoid any saturation of the two-photon fluorescence signal. Also, the excimer laser was operated at similar modest pulse energies and long focal length lens conditions to avoid multiphoton photolysis/excitation effects. Oxygen atom emission at 845 nm was passed through a combination of interference filter and ArF radiation reflector and subsequently detected by a photomultiplier tube.

III. RESULTS

A. H_2/O_2 and H_2/Air Ignition by the ArF Excimer Laser

One of the most important considerations in the development of a practical laser igniter for in-flight use is the laser itself. A tunable laser system, such as is required for O-atom excitation at 226 nm, is not likely to be used in a supersonic aircraft. However, as mentioned before, a much more simple device such as an excimer laser can be envisioned as being made flight worthy. Figure 1 shows the dependence of the incident laser energy (ILE) necessary for the ignition of a premixed flow of H_2/O_2 on the equivalence ratio. The ArF excimer laser was operated in the unstable resonator mode which yields a much less divergent beam as compared to the stable resonator, and thus a tighter focus. The minimum of the curve shows that the ArF laser ignition process is indeed very efficient, with less than 1 mJ pulse energy required. Unlike our previous work using the tunable uv laser, the specific mechanisms for microplasma formation using the broadband (ca. 100 cm^{-1}) fixed frequency of the excimer laser is not yet well-understood. It may include at least two possibilities; (1) a 1+1 multiphoton ionization (MPI) of O_2 involving the Schumann-Runge (S-R) bands, and (2) the 2+1 MPI of H_2 going through the E,F electronically excited states. Determining which of these two mechanisms, or possibly even some other one, is responsible for the efficient ignition awaits further work. For H_2/air, the results are qualitatively similar to those for H_2/O_2 with the exception that the minimum ILE values were found to be around 6 mJ, which is considerably higher than that found for H_2/O_2. We speculate that these ILE values for H_2/air can be dropped considerably by using a tunable excimer laser which is tuned to wavelengths of strong absorption (see below).

B. Microplasma Formation Mechanism

As mentioned above, the microplasma formation mechanism(s) for the ArF excimer laser are not yet fully understood. However, we have conducted further studies of the uv microplasma formation process in general using a tunable laser at 243 nm focussed into a H_2 room temperature flow.

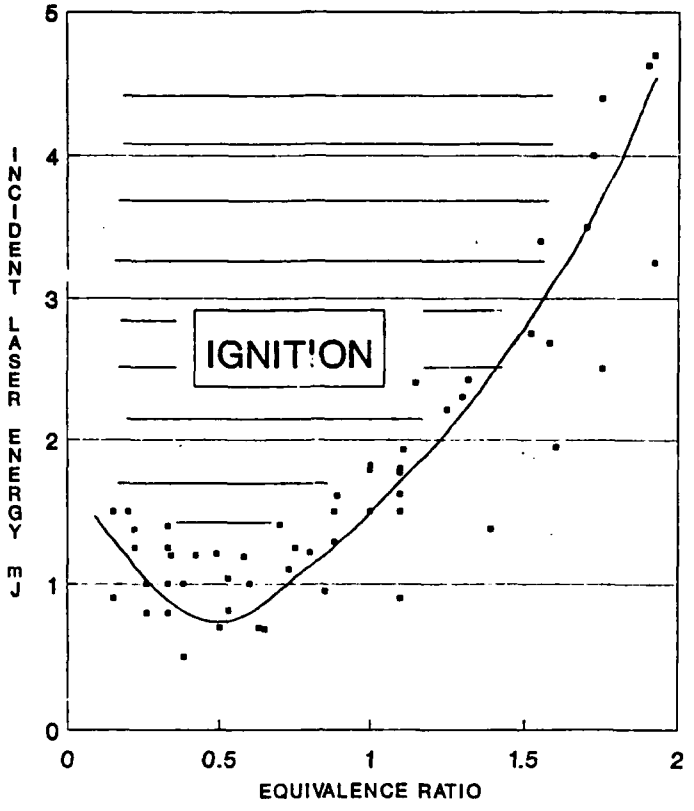

Figure 1. Dependence of ILE on Equivalence Ratio for H_2/O_2 Premixed Flows Using ArF Laser (193 nm) (Unstable Resonator)

Specifically, we compared room temperature D_2 gas behavior with that of H_2 gas. Figure 2 shows the wavelength dependence for microplasma formation in both gases at 70 torr. In both cases we were monitoring the H/D atom emission at 656 nm (n=3 → n=2). A well-defined isotopic shift is clearly evident with a wavelength separation corresponding to about 22 cm^{-1}. This is exactly the energy spacing difference given in energy level tables for the n=2 upper level involved in the two-photon excitation of H and D atoms (n=1 → n=2). We also observe the same isotopic shift at atmospheric pressure with the only difference being broader excitation spectral widths. We believe that these substantial widths observed in ignition/microplasma formation are due to the finite absorption in the "wings" of the atomic transitions. This isotopic shift behavior further substantiates our interpretation of the microplasma formation mechanism, i.e., multiphoton photolysis of parent molecules to form atoms, resonant multiphoton ionization of these atoms, followed by microplasma formation in the laser focal volume using free electrons liberated in the previous step.

Recently we completed a study on the photochemical mechanisms involved in ArF laser photolysis of small carbon-containing molecules.[8] We have expanded this work to include molecular hydrogen. Figure 3 shows the time-of-flight mass spectra (TOF-MS) generated during the irradiation of a molecular beam of H_2 by an ArF (193 nm) excimer laser. Our interpretation of this data is that

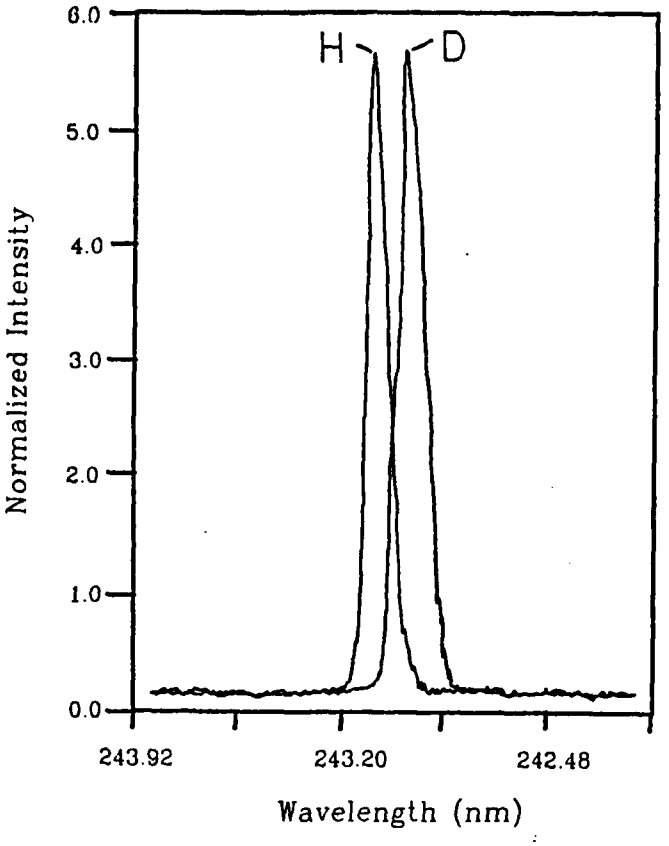

Figure 2. Excitation Spectra for Microplasma Formation in H_2 and D_2 at 70 Torr and Room Temperature. Emission monitored at 656 nm.

under the collisionless conditions of this experiment, the H_2 first ionizes via a 2+1 process involving the E and F states,[9] and then subsequently the molecular ion is photolyzed to produce H ions. If the same experiment is repeated using the laser set at the peak of the two-photon excitation at 243 nm (see Figure 2), then there is no signal from either of these ionic species. Similarly, with the laser set at 225.6 nm (O-atom two-photon transition) we did not detect either the molecular or atomic oxygen ions. This data clearly indicates the importance of collisions in inducing photofragmentation. Studies are currently underway to better understand the importance of collisions on these pathways.

C. Atmospheric Absorption Effects on ArF Laser Ignition Studies

In the course of doing the experiments described in Section A above, we became concerned that the values for the incident laser energy (ILE) that we were measuring may depend on the distance of the ignition site from the laser due to beam attenuation by atmospheric gases, i.e., O_2 absorption in the S-R bands. In order to determine the severity of this potential problem we measured the spectral profile of the transmitted ArF laser beam as propagated through 20 feet of helium gas as compared to 20 feet of air (Figure 4). The He data shows the expected broadband ArF laser spectral profile[10] except for

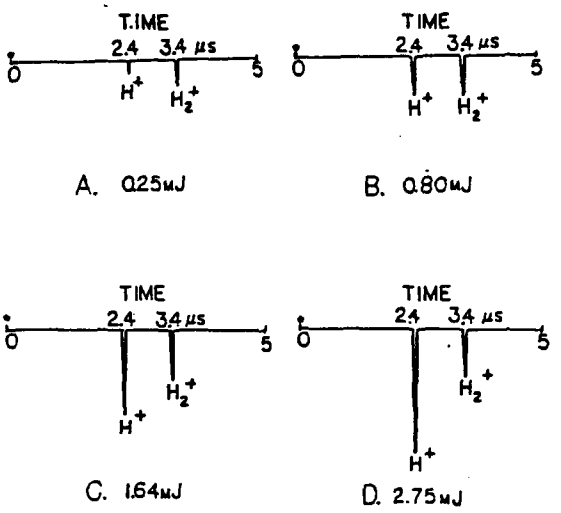

Figure 3. Time-of-Flight Mass Spectra for MPI of H_2 Molecular Beams Using an ArF Excimer Laser (Unstable Resonator). Focussing lens f.l. = 150 mm.

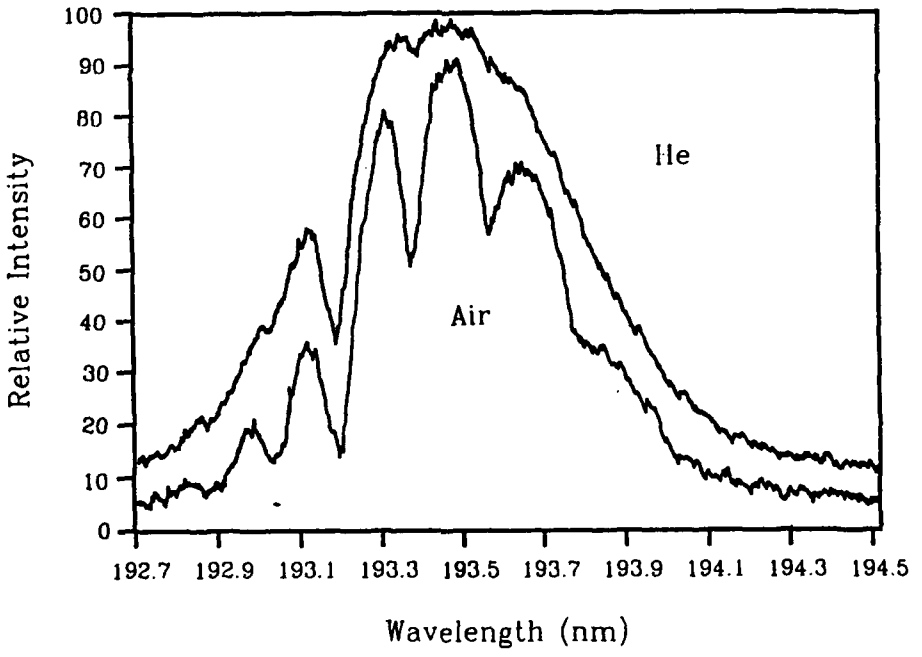

Figure 4. Transmitted Broadband ArF Laser Intensity Through He and Air. Pathlength = 20 feet.

the strong self-absorption feature near 193.1 nm.[11] The air profile, in comparison, clearly shows a number of O_2 absorption line features[10] with the laser beam attenuation measured around 65%. However, the impact of the atmospheric attenuation of the laser beam on the ignition behavior of a premixed H_2/O_2 flow appears to be quite dramatic (Figure 5). The data in Figure 5 suggest that laser radiation within the O_2 absorption spectrum must be important in the ignition process otherwise one would not expect to see such a dramatic difference. Clearly, this phenomenon where laboratory air acts as an "active optical filter" needs to be properly accounted for in ArF laser experiments that are wavelength specific.

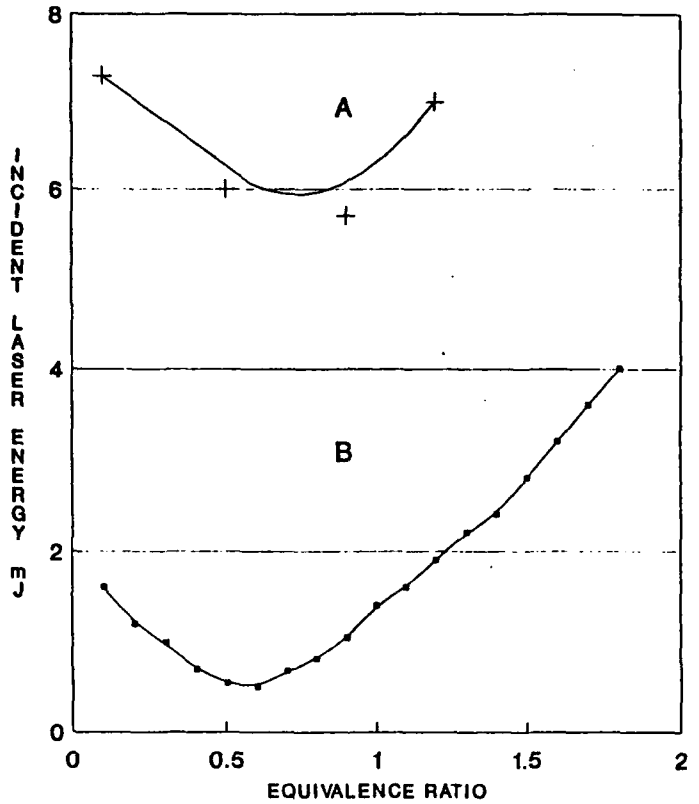

Figure 5. Effect of ArF Laser Attenuation by Laboratory Air on Ignition of H_2/O_2 Premixed Gases.
(A) Pathlength = 20 feet, (B) Pathlength = 1 foot.

D. Nascent Spin-Orbit Distribution of Oxygen Atoms

When photons from an ArF laser beam are absorbed by O_2, the excited molecules predissociate very rapidly such that more than 99% of these excited molecules break apart to form oxygen atoms in the ground electronic state ($2p^4$ ^3P). However, this O-atom state is split into three spin-orbit J states which give rise to the frequently seen spectral "triplet" in fluorescence/ionization

excitation scans or ignition spectral scans around 226 nm.[6] Very little attention has been paid to the nascent distribution of these oxygen atoms into the different spin-orbit states upon photolysis or as reaction products, but this could be important in air-breathing combustion applications, particularly in low pressure/high flow speed conditions where there may not be sufficient time/collisions to "thermalize" these three states. The reason for this is that a substantial difference in the elementary reaction rate constants for the three different O-atom spin-orbit states may exist even for such important combustion reactions as O ($^3P_{2,1,0}$) + H_2 → products. Such spin-orbit state specific rate constant differences have been previously observed in atoms like Br, F, I, Ca, and Sr (typically factors of 2-10) with extreme cases showing 5 orders of magnitude differences.[12] Figure 6 shows a clear case of non-statistical behavior in the photolysis of O_2 by the ArF laser. We have also determined the rate of equilibration for these oxygen-atom spin-orbit states with molecular oxygen as the collision partner (Figure 7).

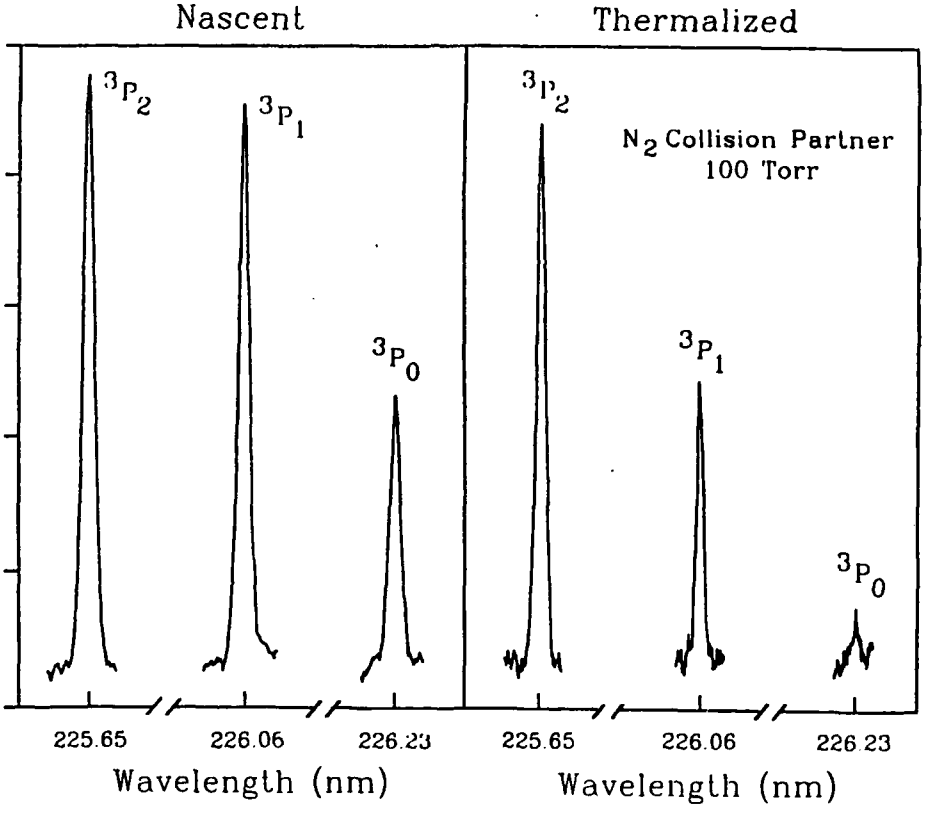

Figure 6. O-Atom Spin-Orbit State Distribution from ArF Laser Photolysis of O_2. Nascent conditions: O_2 = 160 mtorr, 40 nsec delay. Thermalized conditions: O_2 = 300 mtorr, N_2 = 100 torr, 40 nsec delay.

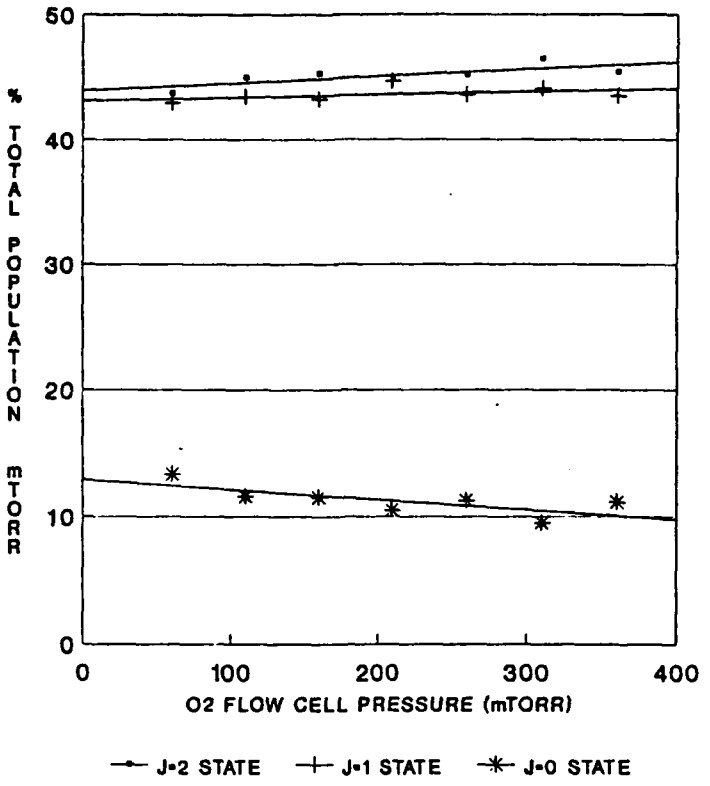

Figure 7. Pressure Dependence for Oxygen-Aton Spin-Orbit Equilibration with O_2 as Collision Partner

E. Ignition of Other Reactive Gases

The study of multiphoton photochemical ignition via fuel molecules was expanded to flowing C_2H_2/air and C_2H_2/O_2 mixtures again irradiated by the ArF (193 nm) excimer laser. Figure 8 shows the time-dependence of the transmitted, ca. 15 nsec, focussed laser beam as it passes above the burner orifice with no flow (Figure 8a) and a C_2H_2 flow (Figure 8b) in which a microplasma is formed. As expected, the bulk of the absorption (Figure 8c) occurs later in the laser pulse, since it takes time for the microplasma (the greatest absorber of radiation) to build-up. Figure 9 shows the dependence of the ILE on equivalence ratio for C_2H_2/air. The scale on the right, i.e., the "upper limit to the minimum ignition energy" was determined by calibrating a laser energy detector which measured the amount of laser energy transmitted with and w/o a reactive flow. The difference represents the amount of laser radiation absorbed and/or scattered. The minimum values around 40 microjoules appear to be a factor of 2 higher than literature values for closed bomb spark ignition of C_2H_2/air.[13] Such a low uv laser ignition energy value, even though higher than the closed bomb spark value, may indicate that the radicals formed near the focal volume might play an important role in ignition kernel growth. Recent experiments using much longer focal length lenses have indicated that the ArF laser can ignite a C_2H_2/air mixture apparently without the need to form a microplasma with, however, somewhat higher levels of ILE required.

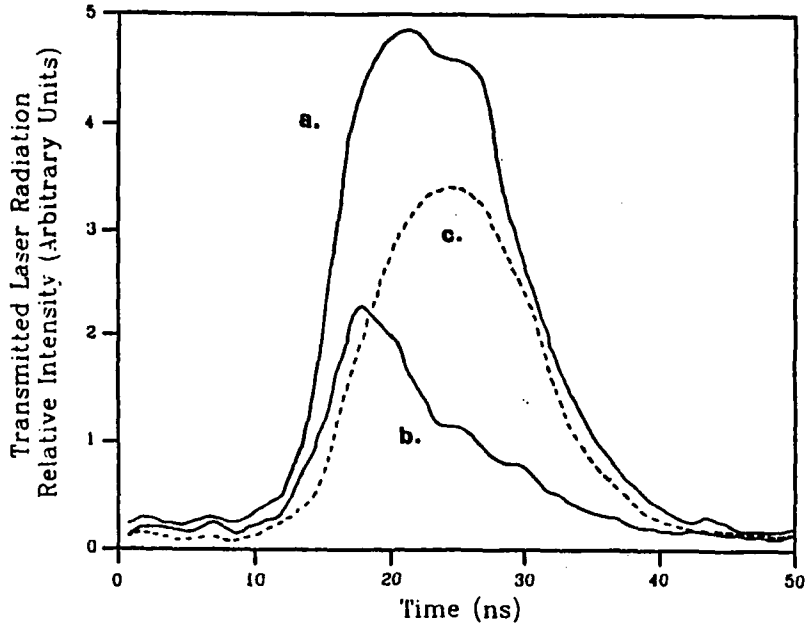

Figure 8. Transmitted ArF Laser Radiation Through a C_2H_2 Flow. (a) C_2H_2 flow absent, (b) C_2H_2 flow present with resulting microplasma, (c) difference between (a) and (b), i.e., absorbed laser energy.

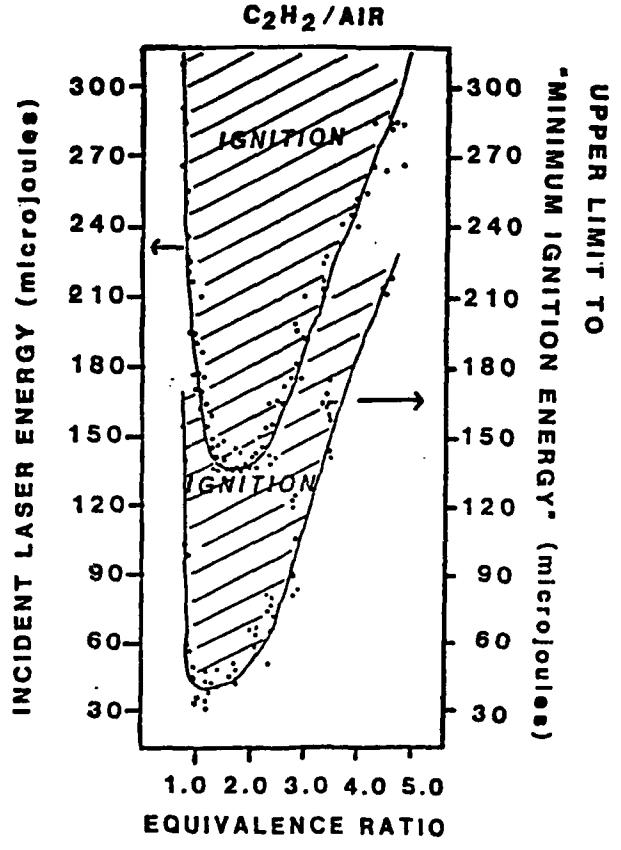

Figure 9. Dependence of ILE on Equivalence Ratio for C_2H_2/Air

IV. SUMMARY

The ArF excimer laser has been demonstrated to readily ignite flows of premixed H_2/O_2, H_2/air, C_2H_2/O_2, and C_2H_2/air. In all cases the laser couples resonantly with one or both of the molecular gaseous constituents. For H_2/O_2 and H_2/air systems the laser energy efficiency would most likely improve if the ArF radiation was tuned to the apropriate molecular (H_2 or O_2) transition. These results are very encouraging with respect to the potential practical application of uv laser ignition for supersonic/hypersonic airbreathing engines. The ArF laser wavelength region (193 nm), however, has certain disadvantages primarily due to atmospheric gas absorption which requires purging of the beam path or use of far-uv optical fibers which are presently quite lossy. Nevertheless, in the case of supersonic/hypersonic reactive flows where the incoming air is substantially shock-heated, laser radiation in the 200-250 nm region may work quite well, but this has not yet been demonstrated.

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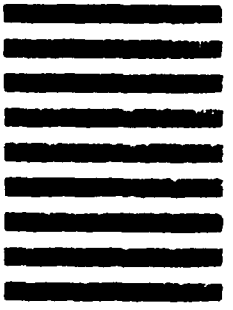

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